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PM-1268

including cocaine from the coca plant and caffeine from coffee beans and cocoa plant products and other xanthine derivatives

PROCESS AND APPARATUS FOR THE SEMICONTINUOUS EXTRACTION OF NICOTINE FROM TOBACCO

Field of the Invention

This invention relates to extraction procedures and is particularly directed to a process for the semicontinuous extraction of materials from plant products. The invention may be used for the selective extraction of particular substances from a great variety of plant products. However, it is particularly applicable and will therefore be described in connection with the extraction of nicotime from tobacco.

Background of the Invention

Various processes have been proposed for the removal of nicotine from tobacco. Most of these processes, however, adversely affect the desirable flavor and aroma properties of the tobacco. Also, they are often complex and expensive to carry out.

United States Patent No. 4,153,063 (Roselius) discloses a process for removing nicotine from tobacco in which tobacco is contacted with an extraction solvent in a supercritical state. It discloses both a single step extraction process and a multi-step extraction process. In the single step extraction process, moist tobacco is extracted with a solvent in a supercritical state. Because

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aroma components are also removed along with nicotine in this single step extraction process, the multi-step process is ~~suggested~~ Preferred. In the first step, dry tobacco is extracted with a solvent in the supercritical state to remove the aroma components. In the second step, the tobacco is moistened and again extracted with a solvent in the supercritical state to remove nicotine. The nicotine is separated from the solvent by either evaporating the solvent, contacting the solvent in a separate vessel with an acid, or adsorbing the nicotine on an active carbon column. In the third step, the stored aroma components from the first step are redissolved in a supercritical solvent and returned to the tobacco. This multi-step extraction process is expensive and time consuming. In addition, the prolonged handling of the aroma components may adversely affect their properties.

Summary of the Invention

This invention provides an improved process for removing nicotine from tobacco. An essentially nicotine-free solvent in the supercritical or liquid state is fed into a first end of an extraction system containing tobacco and a nicotine-rich solvent is discharged from a second end of the extraction system. Periodically a portion of extracted tobacco is discharged from the first end of the extraction flow system while a portion of unextracted tobacco is charged into the second end of the extraction flow system.

This invention also provides an improved process for removing nicotine from an extraction solvent. A nicotine-containing solvent in the supercritical or liquid state is fed into the first end of an entrapment system containing one or more vessels, each vessel containing nicotine entrapment material while an essentially nicotine-free solvent

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is withdrawn from the second end of the entrapment system. Periodically a portion of spent entrapment material is discharged from the first end of the entrapment system while a portion of fresh entrapment material is charged into the second end of the entrapment system.

Persons skilled in the art will recognize that condensation or distillation techniques can also be used to remove nicotine from the solvent and are thus within the scope of this invention.

In one embodiment of this invention, a plurality of tobacco extraction vessels is connected in series as part of a flow system. These extraction vessels are in turn connected to ~~an~~ either one entrapment vessel or a plurality of entrapment vessels which are also connected in series as part of a flow system. Tobacco is extracted with a solvent either in the supercritical state or in the liquid state by continuously passing the essentially nicotine-free solvent through one end of the plurality of extraction vessels connected in series and discharging the nicotine-enriched solvent from the opposite end. Thereafter the solvent, enriched in nicotine, is passed through an entrapment vessel or a plurality of entrapment vessels, connected in series, to remove the nicotine. The solvent, depleted in nicotine, is then recycled to the extraction vessel or vessels to again extract nicotine. Removal and addition of an extraction vessel from the flow system, or removal and addition of an entrapment vessel from the flow system, to provide continuous extraction or entrapment, respectively, is accomplished by valve adjustment.

It is an object of this invention to provide a process for extracting nicotine from tobacco which is more efficient, provides a faster cycle time and results in lower capital and operating costs, than earlier processes

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It is another object of this invention to provide a process for extracting nicotine from tobacco which increases the concentration of nicotine in the solvent and decreases the amount of solvent required per unit of tobacco.

It is still another object of this invention to provide a process for extracting nicotine from tobacco which requires less extraction solvent and thereby results in less degradation and loss of the aroma producing components and consequently gives an improved tobacco product.

It is another object of this invention to provide a process for extracting nicotine from tobacco which increases the amount of nicotine loaded on the nicotine entrapment material and significantly decreases the ratio of entrapment material to tobacco.

It is a further object of this invention to provide a process for extracting nicotine from tobacco which results in a reduced CO_2 pressure drop, reduced tobacco bed compaction and a more favorable extraction bed geometry.

These and other objects and advantages of the invention may be seen in the following description.

Brief Description of the Drawing

FIG. 1 illustrates an apparatus for the semi-continuous extraction of nicotine from tobacco.

FIG. 2 illustrates an apparatus for the semi-continuous extraction of nicotine from tobacco and the semi-continuous entrapment of nicotine from the solvent.

Detailed Description of the Invention

An apparatus for the semi-continuous extraction of nicotine from tobacco is shown in FIG. 1. Extraction vessels 10 through 14 can contain tobacco

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and can be connected in series as part of a flow system by valve adjustment. The vessels 10 and 11 as depicted are on stream and are connected to entrapment vessel 15.

The extraction vessels are preferably all designed for radial flow or all for axial flow of solvent. The entrapment vessel can be designed for radial flow or axial flow. A radial flow of solvent will minimize compaction of solid material in a vessel and may allow for lower pressure drops within each vessel. Persons skilled in the art will recognize that many directions of flow will be effective, e.g., flow from bottom to top or top to bottom or inward or outward radially in each vessel. Persons skilled in the art will also recognize that the pump can be placed on any of several lines in the system.

In a preferred form of the invention, an extraction solvent is ~~supplied~~ circulated through pump 16 to extraction vessel 10 ~~which is connected to pump 16~~. The pressure in the vessel is controlled by means of a fill pump (not shown) and the temperature is controlled by means of heat exchanger 17. The extraction solvent enters the top of extraction vessel 10, passes downwardly through the tobacco bed and leaves at the bottom of the vessel. In passing through the tobacco bed, the extraction solvent becomes enriched with nicotine from the tobacco. The solvent is then circulated to extraction vessel 11, again being introduced from the top, and then passing downwardly and exiting at the bottom. After exiting extraction vessel 11, the solvent is circulated to entrapment vessel 15. The extraction solvent enters the top of the vessel and then passes downwardly exiting at the bottom. In passing through the vessel, the nicotine in the solvent becomes trapped on the entrapment material. The solvent, depleted of nicotine, is then returned or in

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into the cycle by recirculating it to extraction vessel 10.

Extraction vessels 12, 13 and 14 are off stream and are in the turn-around cycle. In the turn-around cycle, extraction solvent is vented from the extraction vessel, the extracted tobacco is unloaded, unextracted tobacco is loaded into the extraction vessel, and the extraction vessel is refilled with extraction solvent. Extraction vessels containing extracted tobacco are removed periodically from the end of the extraction flow system into which essentially nicotine-free solvent is fed while simultaneously extraction vessels containing unextracted tobacco are added at the end of the extraction flow system from which nicotine-enriched solvent is discharged. Removal and addition of extraction vessels from or to the flow system is accomplished by valve adjustment.

FIG. 2 illustrates an alternative, preferred embodiment wherein a plurality of extraction vessels, connected in series, is connected ~~with~~ to a plurality of entrapment vessels, also connected in series. Extraction vessels 10 and 11 contain tobacco and are connected in series and are on stream. Extraction vessels 12, 13 and 14 are off stream and in the turn-around cycle. Entrapment vessels 20 and 21 are connected in series and are on stream. Entrapment vessel 22 is off stream and in the turn-around cycle.

The extraction vessels are preferably all designed for radial flow or all for axial flow of solvent. The entrapment vessels are also preferrably all designed for radial flow or axial flow but need not be of the same design as the extraction vessels. A radial flow of solvent will minimize compaction of solid material in a vessel and may allow for lower pressure drops within each vessel. Persons skilled in the art will recognize that many directions of

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flow will be effective, e.g., flow from bottom to top or top to bottom or inward or outward radially in each vessel. ~~Persons skilled in the art will also recognize that the pump can be placed on any of several lines in the system.~~

As described for FIG. 1, extraction solvent is supplied to extraction vessel 10 and then circulated to extraction vessel 11. After exiting extraction vessel 11, the solvent is circulated to entrapment vessel 20. The extraction solvent enters the top of the vessel and passes downwardly, exiting at the bottom. The solvent is then circulated to entrapment vessel 21 again being introduced from the top, and passing downwardly, exiting at the bottom. In passing through the vessels 20 and 21, the nicotine in the solvent becomes trapped on the entrapment material. The solvent, essentially depleted of nicotine, is then returned into the cycle by recirculating it to extraction vessel 10.

Entrapment vessel 22 is off stream and in the turn-around cycle. In the turn-around cycle, extraction solvent is vented from the entrapment vessel, the spent entrapment material unloaded, fresh entrapment material is loaded into the vessel and the vessel is refilled with extraction solvent. Entrapment vessels containing spent entrapment material are removed periodically from the end of the entrapment flow system into which nicotine-enriched solvent is fed while simultaneously entrapment vessels containing fresh entrapment material are added at the end from which nicotine-lean solvent is discharged. Removal and addition of entrapment vessels from or to the flow system is accomplished by valve adjustment.

In yet another embodiment of this invention, a plurality of entrapment vessels connected in series may be used to remove nicotine from a solvent in a process utilizing a single extraction vessel rather

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than a plurality of extraction vessels connected in series.

A number of extraction solvents having solvent capacity for nicotine in both their liquid and gaseous phases can be employed to reduce the nicotine content of tobacco. Carbon dioxide in the supercritical state is the preferred solvent in this invention. Other useful solvents include, for example, halogenated hydrocarbons including up to about 4 carbon atoms such as CF_4, CHF_3CClF_3, $CBrF_3$, $CF_2=CH_2$, $CF_3-CF_2CF_3$, $CHClF_2$, CCl_2F_2, $CHCl_2F$, CCl_3F, $CBrF_3$, $CFCl=CF_2$, CH_3-CF_3, octafluorocyclobutane and hydrocarbons including up to about 5 carbon atoms such as propane, butane, pentane; other useful solvents include N_2O, SF_6 and argon. Mixtures of solvents or additives or co-solvents may be used to obtain improved solvent characteristics.

A solvent in the supercritical state is a solvent in the gas phase at a sufficiently high temperature so that it cannot be liquefied by an increase in pressure. A solvent in the subcritical state is a solvent which can be liquefied by an increase in pressure.

Supercritical carbon dioxide is carbon dioxide which is above its critical temperature, i.e., above about 31.3°C. and above its critical pressure, i.e., above about 70 atmospheres. Extraction with carbon dioxide in the supercritical state is carried out at a pressure in the range of from about 70 to about 1500 amospheres and at a temperature in the range of from above about the critical temperature to about 120°C. The range of temperature and pressure for the supercritical state of other useful solvents are of generally the same order of magnitude.

The entrapment material in the entrapment vessel may be an adsorbent or absorbent with an

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affinity for nicotine. ~~Useful~~ adsorbents include activated carbon, silica, alumina, magnesium silicate and ion exchange resins. The adsorbent may also be mixed with a diatomaceous earth, up to a ratio of about 1:1, to improve the flow rate of the adsorbent. Other useful absorbents include tobacco or tobacco stems, tobacco plant products which have been treated with an acid, and other absorbents such as cocoa shells. One particularly useful absorbent is tobacco stems which have been sprayed with or soaked in an aqueous solution of a polycarboxylic acid or salts thereof, such as monopotassium citrate.

Alternatively, the entrapment material in the entrapment vessel may be an absorbent which has an affinity for nicotine. Absorbents are preferred over adsorbents. Such absorbents include water, acid, aqueous acid solutions and aqueous salt solutions.

The preferred acids for use as an entrapment material in this invention are non-volatile and non-soluble in the solvent under the conditions of the extraction. ~~Useful~~ acids ~~are~~ sulfuric, phosphoric and nitric. Other useful acids ~~are~~ polycarboxylic acids such as tartaric, citric, malic, lactic, malonic, succinic, acetic and glutamic.

Monovalent salts such as the alkali salts of the above acids are generally preferred because these salts are less volatile and less soluble in the solvent. A preferred salt of an acid is monopotassium citrate. Monoammonium and diammonium salts of the above acids may also be used. Polyvalent salts of the above acids are also useful but are less efficient in trapping nicotine.

The extraction process may be carried out on tobacco which has or has not been premoistened. It is generally preferred to moisten the tobacco up to about 25% OV (oven volatiles). The percentage of oven volatiles (% OV) in the tobacco is a measure of

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the moisture content plus a minor fraction of other components and is determined as follows:

$$\% \text{ OV} = \frac{\text{weight loss of sample after 3 hrs. at 100°C}}{\text{sample weight}}$$

In a typical semi-continuous extraction and entrapment, as illustrated in FIG. 2, the cycle time for an extraction vessel is 1 hour and the total extraction is 2 hours. Turn-around time for the extraction vessel is 3 hours. Cycle time for an entrapment vessel is 3 hours and the total entrapment (adsorption/absorption) time is 6 hours. Turn-around time for an entrapment vessel is 3 hours.

Table 1 shows a comparison of nicotine concentration in supercritical carbon dioxide in a batch extraction versus a semicontinuous extraction. The amount of nicotine removed from the tobacco is shown, on a dry weight basis (dwb). The average nicotine concentration in the carbon dioxide is lower in a batch extraction versus a semi-continuous extraction which results in a decrease in the carbon dioxide/tobacco ratio from (300:1 to 168:1.)

Table 1

COMPARISON OF
BATCH vs. SEMICONTINUOUS EXTRACTION

EXTRACTION TIME (hrs)	NICOTINE IN TOBACCO (%)	NICOTINE REMOVED, (dwb) (g/100g)	NICOTINE CONC. IN CO_2 (ppm)	
			SEMI-	
			BATCH	CONTINUOUS
0	100	0		
1.0	86.8	1.605	107	107
2.0	2.7	0.195	13	107
		1.80	60	107
		(total)	(avg)	(avg)

Table 2 shows a comparison of the carbon to tobacco ratio, on a dry weight basis (dwb), in a

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batch extraction-entrapment, a batch extraction-semicontinuous entrapment and a semicontinuous extraction-entrapment. The carbon to tobacco ratio dropped from 4.0 in a batch extraction-entrapment to about 2.0 in a semi-continuous entrapment, which represents a decrease of 50% in the carbon to tobacco ratio required. The estimated carbon to tobacco ratio in the semi-continuous extraction and entrapment is significantly below 2.0 (in the range of 0.4 to 0.8) which even when calculated based on 0.8 still represents a decrease of 80% in the carbon to tobacco ratio when compared versus a batch extraction-entrapment and a decrease of 60% when compared with the semi-continuous extraction.

EXAMPLE 1

Batch Extraction/Batch Entrapment

This experiment example illustrates old previous technology in order to better show the advantages of the present invention.

American blend tobacco, 5.5 Kg dwb was moistened to 25% OV via water spray in a rotary cylinder. The moistened tobacco was loaded into an extractor vessel. 22 Kg of activated carbon was loaded into an entrapment vessel. The system was pressurized with CO_2 to 260 bar and 70°C. The flow of CO_2 was maintained from the tobacco extraction vessel to the carbon entrapment vessel for 60 minutes. Then, CO_2 was vented and tobacco and carbon were unloaded. A profile of nicotine concentration on carbon in the entrapment vessel indicated that all of the carbon was needed for nicotine entrapment, i.e., a low level of nicotine was present at the outlet of the adsorber. About 97% of nicotine in the tobacco was removed. Run details were:

Wt. Tobacco	=	5.5 Kg dwb
Wt. Carbon	=	22 Kg
Carbon/Tobacco Ratio	=	4/1

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CO_2 Flow = 1.1 m^3/hr

$M_{CO_2}/M_{Tobacco}$ = 150

EXAMPLE 2

Batch Extraction/Semicontinuous Entrapment

Simulation of a semicontinuous nicotine entrapment was done, as follows.

American blend tobacco, 5.5 Kg dwb, was moistened to 25% OV via water spray in a rotary cylinder. The moistened tobacco was loaded into an extractor vessel. 11 Kg of activated carbon was loaded into each of two entrapment vessels. The system was pressurized to 260 bar and 70°C with CO_2. CO_2 flow was maintained from tobacco to one carbon vessel for 30 minutes and to the second entrapment vessel for another 30 minutes. The total extraction time was 1.0 hr. At the end of the extraction, CO_2 was vented, and carbon in Adsorber (entrapment vessel) 1 was replaced with fresh activated carbon. A fresh batch of tobacco was loaded into the extractor, but the carbon in Adsorber 2 was not unloaded in order to reuse it. Supercritical CO_2 at 260 bar and 70°C was cycled through the fresh batch of tobacco ~~was extracted.~~ Adsorber 2 ~~for entrapment of nicotine~~ for ~~the first~~ 30 minutes and then through Adsorber 1 for ~~the last~~ 30 minutes. This continuous entrapment was continued for six batches of fresh tobacco. Nicotine extraction level of 96.5% or better was achieved for each batch of tobacco.

Run details were:

Wt. Tobacco = 5.5 Kg dwb

Wt. Carbon = 22 Kg total, (11 Kg ~~effective~~ per cycle)

~~Wt. Carbon~~

Ratio Carbon/Tobacco = 2/1 (effective)

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CO_2 Flow = 1.1 m^3/hr

$M_{CO_2}/M_{Tobacco}$ = 150

Run	Tobacco Wt., Kg. dwb	Extraction Time (Min.) Ads 1	Ads 2	Total	Nicotine Removal %
1	5.5	30	30	60	97.2
2	5.5	30	30	60	96.5
3	5.5	30	30	60	97.0
4	5.5	30	30	60	97.7
5	5.5	30	30	60	97.0
6	5.5	30	30	60	97.2

Note: Carbon in second adsorber was reused in the next batch as the first adsorber.

TABLE 2

~~Summary of Examples 1 and 2~~

	Carbon/Tobacco Ratio (dwb)
Batch Extraction/ Batch Entrapment	4
Batch Extraction/ Semicontinuous Entrapment	about 2
Semicontinuous Extraction/ Semicontinuous Entrapment	less than 2[1] (~~0.4 - 0.8 estimated~~)

1. Projected ratio - estimated to be 0.4 - 0.8 in practice

Table 3 shows a comparison of design features of a batch extraction and a semi-continuous extraction and entrapment. Full flavor tobacco having a nicotine content of 1.85% (dwb) and an oven volatile content of 25% is extracted with carbon dioxide under supercritical conditions at 260 atmospheres and 70°C using carbon as the entrapment material. After extraction, the tobacco has a nicotine content of 0.05%.

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The number and size of the vessels required in a semi-continuous system are smaller than in a batch system resulting in reduced turn-around time. In addition, the number of CO_2 circulation pumps, the size of the CO_2 storage vessels and the size of the CO_2 handling system (fill pump, compressor) are smaller. Only one heat exchanger, one recovery cooler-condensor and one dust filter are needed. The average nicotine concentration in the super-critical CO_2 is higher, which results in a lower value for the CO_2 to tobacco ratio and a lower value for the entrapment material to tobacco ratio. In addition, because the system runs semi-continuously, equipment reliability is better and adaptability of the system to a liquid wash process is ~~easier~~ improved.

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Table 3

		BATCH EXTRACTION/ SEMICONTINUOUS ENTRAPMENT (4 UNITS)	SEMI-CONTINUOUS EXTRACTION/ ENTRAPMENT (4 UNIT EQUIV.)
	CYCLE TIME	5 hrs	1 hr. extraction, 3 hrs. adsorption
	EXTRACTION TIME	2 hrs	2 hrs
	TURN-AROUND TIME	3 hrs	3 hrs
	$^{M}CO_2/^{M}TOB.$ RATIO[1]	300	168
	CARBON/TOB. RATIO	2	≤ 2
	CO_2 FLOW	$0.681(\bar{M})$ lbs/hr[2] (per unit or 2.724 $\bar{M}$ lbs/hr total for 4 units)	$0.611(\bar{M})$ lbs/hr (total)
	EXTRACTION VESSELS:		
	NO. OF VESSELS	4	5
	TOTAL TOB. CHARGE, LBS. (dwb)	18,168	18,168
	PER VESSEL TOB. CHARGE, LBS. (dwb)	4,542	3,634
	PER VESSEL TOB. VOLUME, FT^3	458	366
	NOMINAL VESSEL VOLUME, FT^3	2,765	640
	NOMINAL VOLUME SIZE, FT	8D X 55H	4.5D X 40H
	ENTRAPMENT VESSELS:		
	NO. OF VESSELS	8	3
	TOTAL CARBON CHARGE, LBS.	72,672	<65,600
	PER VESSEL CARBON CHARGE, LBS.	9,084	<21,870
	PER VESSEL CARBON VOLUME, FT^3	364	< 875
	NOMINAL VESSEL VOLUME, FT^3	804	TBD
	NICOTINE:		
	LOADING ON CARBON, %	0.9	0.9+
	LBS. REMOVED/CHARGE	328	65.6
	CO_2 FLOW($\bar{M}$ LBS/HR)	2.724	0.611
	AVG. NICOTINE CONC. IN CO_2	60	107
	$^{M}CO_2/^{M}TOB.$ RATIO[1]	300	168
	TOTAL NO. OF PRESS. VESSELS	12	8

1 $^{M}CO_2/^{M}TOB.$ = mass of CO_2/mass of tobacco

2 $\bar{M}$ lbs/hr = million pounds per hour

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EXAMPLE 3

Semicontinuous Extraction/Semicontinuous Entrapment

The following example is illustrative.

Eight pressure vessels are arranged for series extraction as shown in FIG. 2. The extraction vessels (10 through 14) are large enough to hold 4,845 lbs. of tobacco each at 25% OV (i.e., 366 cu.ft. of tobacco). The entrapment vessels (20 through 22) are large enough to hold 21,870 lbs. of activated carbon (i.e., 875 cu.ft. of carbon volume).

Dry carbon (21,870 lbs.) is loaded into each entrapment vessel (20, 21, 22). A blend of full flavor American tobacco is moistened from about 12% OV to about 25% OV by direct spray of deionized water in a rotating cylinder. About 4,845 lbs. of premoistened tobacco (i.e., 3,634 lbs. dwb) is loaded into each extraction vessel (10, 11, 12, 13, 14).

Start-Up

A CO_2 fill pump is used to pressurize vessels 10 and 20. Carbon dioxide is circulated through the ~~two vessels~~ flow system at the rate of 611,000 lbs/hr. After extraction conditions are reached, i.e., 260 atmospheres, 70°C, the circulation of CO_2 is continued through vessels 10 and 20 for one hour. Vessels 11 and 21 are pressurized with CO_2 during this first hour.

In the second hour of extraction, the flow of CO_2 is directed through vessels 10, 11, 20 and 21 as shown in FIG. 2. Vessel 12 is pressurized with CO_2 during this second hour.

In the third hour of operation, the flow of CO_2 is directed through vessels 11, 12, 20 and 21. ~~Also in the third hour of operation, extraction vessel 10 is removed from the extraction loop and subjected to the turn-around phase.~~ The CO_2 in vessel 10 is vented and the extracted tobacco is unloaded. Vessel 10 is again filled with tobacco and CO_2 and is ready to be returned to the extraction

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~~for~~ and prepared for

During the turn-around phase,

Vessel 22 is pressurized with CO_2 during this third hour, and turn-around of vessel 10 is begun.

~~To~~ extraction vessel 10 is removed from and vessel 12 is added to the flow system.

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loop. The turn-around phase for an extraction vessel takes 3 hours.

In the fourth hour of operation, vessel 13 is added to the extraction loop and vessel 11 is removed from the turn-around phase.

Also in the fourth hour of operation, the flow of CO_2 is directed through entrapment vessels 21 and 22. Entrapment vessel 20 is subjected to the turn-around phase. The CO_2 in vessel 20 is vented and the spent carbon is unloaded. Vessel 20 is again filled with carbon and CO_2 and is ready to be returned into the extraction loop. The turn-around phase for an entrapment vessel takes 3 hours.

Steady state conditions are reached in six hours. Vessel 10 is returned to the extraction loop in the 6th hour of operation (Table 4). Extraction vessels 11, 12, 13 and 14 are each similarly subjected to the turn-around phase and then returned to the extraction loop.

Continuous Operation

As shown in FIG. 2 and Table 4, one batch of extracted tobacco is produced every hour. The nicotine content in the tobacco is reduced 97%, from 1.85% nicotine (dwb) to 0.05% nicotine (dwb). The total extraction time is two hours. The solvent to tobacco ratio is 168 parts of CO_2 to one part of tobacco (dwb). This solvent to tobacco ratio is significantly lower than the 300 parts of CO_2 to one part of tobacco (dwb) required in a batch system.

As shown in Table 5, it is found that the continuous operation increases the concentration of nicotine in the CO_2 solvent without significantly increasing the concentration of other tobacco soluble materials (considered to be important for product quality) in the CO_2 solvent.

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Table 4

	Time (Hr.)	CO_2 Flow Arrangement (1) Extraction Vessels	Entrapment Vessels	Vessels in Turn-around Phase (2)	
Start-up:	1	10	20		
	2	10,11	20,21		
	3	11,12	20,21	10	
	4	12,13	21,22	10,11,20	
	5	13,14	21,22	10,11,12,20	
	6	14,10	21,22		11,12,13,20
Continuous Steady State Operation:	7	10,11	22,20	12,13,14,21	
	8	11,12	22,20	13,14,10,21	
	9	12,13	22,20		14,10,11,21
	10	13,14	20,21	10,11,12,22	
	11	14,10	20,21	11,12,13,22	
	12	10,11	20,21	12,13,14,22	
	13	11,12	21,22	13,14,10,20	
	14	12,13	21,22		14,10,11,20
	15	----------Continous Operation----------			

Note:

1 - CO_2 flow maintained at 611,000 lb/hr through vessels indicated.

2 - Turn-around time is 3 hours. Turn-around time (TAT) includes the following steps:

CO_2 vent

Unload ~~Tobacco or activated carbon unloading~~

Load ~~Tobacco or activated carbon loading~~

Pressurize CO_2 ~~fill~~ to extraction conditions (260 atmospheres, 70°C)

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Table 5

	Batch Operation	Continuous Operation
% Nicotine in Tobacco (dwb)		
Unextracted	1.85	1.85
Extracted	0.05	0.05
Solvent/Tobacco Ratio (dwb)	300	168
Solubles in CO_2 (ppm)		
Nicotine	60	107
Other Tobacco Solubles	60-120	60-120
% Solubles Removed from Tobacco (dwb)		
Nicotine	1.8	1.8
Other Solubles	1.8-3.6	1.0-2.0

Expert evaluation shows that cigarettes made from extracted tobacco, where the solvent to tobacco ratio is low, are of higher subjective quality than cigarettes made from extracted tobacco where the solvent to tobacco ratio is high.

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I claim:

1. A method for the extraction of nicotine from tobacco which comprises:

(a) feeding an essentially nicotine-free solvent to a first end of an extraction flow system containing tobacco and withdrawing a nicotine-rich solvent from a second end of the extraction flow system;

(b) periodically discharging a portion of extracted tobacco from the first end of the extraction flow system; and

(c) charging a portion of unextracted tobacco to the second end of the extraction flow system.

3 2. The method according to claim 1 wherein the solvent is in the supercritical state.

4 3. The method according to claim 1 wherein the solvent is in the liquid state.

2 4. The method according to claim 1 wherein the moisture content of the tobacco is to about 30% by weight.

5. The method according to claim 1 wherein the extraction solvent is selected from the group consisting of carbon dioxide, argon, SF_6, N_2O, a lower hydrocarbon and a lower halogenated hydrocarbon.

6. The method according to claim 5 wherein the extraction solvent is carbon dioxide.

7. The method according to claim 1 wherein the extraction process is carried out in a plurality of extraction vessels connected in series.

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8. A method of separating nicotine from a solvent which comprises:

(a) feeding a nicotine-containing solvent to a first end of an entrapment vessel containing a nicotine entrapment material and withdrawing an essentially nicotine-free solvent from a second end of the entrapment flow system;

(b) periodically discharging a portion of spent entrapment material from the first end of the entrapment flow system; and

(c) charging a portion of fresh entrapment material to the second end of the entrapment flow system.

9. The method according to claim 8 wherein the solvent is in the supercritical state.

10. The method according to claim 8 wherein the solvent is in the liquid state.

11. The method according to claim 8 wherein the extraction solvent is selected from the group consisting of carbon dioxide, argon, SF_6, N_2O, a lower hydrocarbon and a lower halogenated hydrocarbon.

12. The method according to claim 11 wherein the extraction solvent is carbon dioxide.

13. The method according to claim 8 wherein the entrapment material is an adsorbent selected from the group consisting of carbon, silicon, alumina, magnesium silicate and ion exchange resins.

14. The method according to claim 8 wherein the entrapment material is an absorbent selected from the group consisting of water, acid, aqueous acid and aqueous salt solutions.

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15. The method according to claim 14 wherein the entrapment material is an aqueous solution containing monopotassium citrate.

16. The method of according to claim ~~14~~ 8 wherein the entrapment material is tobacco stems which have been pretreated with monopotassium citrate.

17. The method according to claim 8 wherein the entrapment process is carried out in a plurality of entrapment vessels connected in series.

18. A method for the extraction of nicotine from tobacco which comprises:

(a) feeding an essentially nicotine-free solvent to a first end of an extraction flow system containing tobacco and withdrawing a nicotine-containing solvent from a second end of the extraction flow system;

(b) periodically discharging a portion of extracted tobacco from the first end of the extraction flow system;

(c) charging a portion of unextracted tobacco to the second end of the extraction flow system;

(d) feeding a nicotine-containing solvent to the first end of an entrapment flow system containing a nicotine entrapment material and withdrawing an essentially nicotine-free solvent from the second end of the entrapment flow system;

(e) periodically discharging a portion of spent entrapment material from the first end of the entrapment flow system; and

(f) charging a portion of fresh entrapment material to the second end of the entrapment flow system.

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19. The method according to claim 18 wherein the moisture content of the tobacco is up to about 30% by weight.

20. The method accoding to claim 18 wherein the solvent is in the supercritical state.

21. The method according to claim 18 wherein the solvent is in the liquid state.

22. The method according to claim 18 wherein the solvent is selected from the group consisting of carbon dioxide, argon, SF_6, N_2O, a lower hydrocarbon and a lower halogenated hydrocarbon.

23. The method according to claim 22 wherein the solvent is carbon dioxide.

25 ~~24.~~ The method according to claim 18 wherein the entrapment material is selected from the group consisting of carbon, silicon, alumina, magnesium silicate and ion exchange resins.

26 ~~25.~~ The method according to claim 18 wherein the entrapment material is selected from the group consisting of water, acid, aqueous acid and acqueous salt solutions.

~~26.~~ The method according to claim 25 wherein the entrapment material is an aqueous solution ~~of~~ containing monopotassium citrate.

26 ~~27.~~ The method according to claim 26 wherein the entrapment material is an aqueous solution containing monopotassium citrate.

24. The method according to claim 22 wherein the extraction process is carried out in a plurality of extraction vessels connected in series.

27. The method according to claim ~~22~~ 18 wherein the entrapment material is tobacco stems which have been pretreated with monopotassium citrate

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27 Met 28. The method according to claim 18 wherein the extraction process is carried out in a plurality of extraction vessels connected in series.

29. The method according to claim 18 wherein the entrapment process is carried out in a plurality of entrapment vessels connected in series.

30. An apparatus for the extraction of nicotine from tobacco with a solvent in the supercritical state or in the liquid state which comprises a plurality of extraction vessels connected in series.

31. The apparatus according to claim 30 wherein the extraction solvent is carbon dioxide.

32. An apparatus for the removal of nicotine from a solvent in the supercritical state or in the liquid state which comprises a plurality of entrapment vessels connected in series.

33. The apparatus according to claim 32 wherein the extraction solvent is carbon dioxide.

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PROCESS AND APPARATUS FOR THE SEMICONTINUOUS EXTRACTION OF NICOTINE FROM TOBACCO

Abstract of the Disclosure

A process is provided for the improved removal of nicotine from tobacco. An essentially nicotine-free solvent in the supercritical or liquid state is fed into a first end of an extraction flow system containing tobacco and a nicotine-rich solvent is discharged from a second end of the extraction flow system. Periodically a portion of extracted tobacco is discharged from the first end of the extraction system while simultaneously a portion of an unextracted tobacco is charged to the second end of the extraction system. Nicotine is then entrapped or otherwise removed from the solvent and solvent is recycled through the extraction flow system.

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PM-1268

DECLARATION AND POWER OF ATTORNEY

We, Ravi Prasad and Harvey J. Grubbs, declare that we are citizens of the United States of America, residing and having post office addresses, respectively, at 108-21 Henshaw Drive, Midlothian, Virginia 23110 and Route 4, Box 276K, Mechanicsville, Virginia 23111;

that we verily believe ourselves to be the original, first and sole inventors of the invention or discovery in:

PROCESS AND APPARATUS FOR THE SEMICONTINUOUS EXTRACTION OF NICOTINE FROM TOBACCO

which is described and claimed in the attached specification;

that we have read and do understand the content of said specification, including the claims, and knowledge and duty to disclose information, of which we are aware, which is material to the examination of this application in accordance with Title 37, Code of Federal Regulations, § 1.56(a);

that we do not know and do not believe that this invention or discovery was ever known or used in the United States of America before our invention or discovery thereof, or patented or described in any printed publication in any country before my invention or discovery thereof, or more than one year prior to this application; or in public use or on sale in the United States of America more than one year prior to this application; that this invention or discovery has not been patented or made the subject of an inventor's certificate issued before the date of this application in any country foreign to the United States of America on an application filed by us or our legal representatives or assigns

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more than twelve months prior to this application; and that no application for patent or inventor's certificate on this invention or discovery has been filed in any country foreign to the United States of America by us or our legal representatives or assigns;

and we hereby appoint Arthur I. Palmer, Jr., Reg. No. 18,855, James E. Schardt, Reg. No. 23,033, Albert E. Fey, Reg. No. 19,018, W. Edward Bailey, Reg. No. 30,994, and Thomas L. Secrest, Reg. No. 30,145, our attorneys, with power of substitution, and with power of appointment of associate attorneys, and of revocation of their powers, to prosecute this application and any divisions, continuations in whole or in part, renewals and reissues of the same, and to transact all business in the Patent and Trademark Office connected therewith;

and we request that communications be sent to:

Thomas L. Secrest
c/o Fish & Neave
875 Third Avenue
New York, New York 10022-6250

and that telephone calls be directed to:

Thomas L. Secrest
(212) 715-0600.

Wherefore, we pray that Letters Patent be granted to us for the invention or discovery described and claimed in the attached specification and claims, and we hereby subscribe our names to the foregoing specification and claims, declaration, and power of attorney.

We declare, further, that we understand the English language and that all statements made herein of our own knowledge are true, and that all statements made on information and belief are believed to be true; and, further, that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or

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imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Date	Ravi Prasad
Date	Harvey J. Grubbs

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